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An Overview of Microelectronic Infrared Pyroelectric Detector

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Abstract

Pyroelectric materials can operate with a high thermodynamic efficiency, compared to thermoelectric, it does not require bulky heat sinks to maintain the temperature gradient. Many materials that can be used for the production of infrared pyroelectric detectors exist. The choice of one is a difficult task because of many parameters involved in the calculation of the desired characteristics of the device that has to be designed, such as detector size, operating temperature, frequency of operation, and *etc*. A material of a lower performance than triglycine sulphate (TGS), but possessing high chemical stability, low loss, good figure of merit, high Curie temperature and good insolubility in water is the lithium tantalite. In this review, the pyroelectric properties of various materials are discussed how they can improve the properties of detector in potential applications.

Keywords: Infrared detectors; Pyroelectric materials; Pyroelectric sensor; Pyroelectric effect; Pyroelectric co-efficients.
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1. Introduction:

Researchers have developed great interest in the detection of infrared radiation for a wide range of applications, such as optical gas sensing in the infrared spectral range.[1] Nowadays, there is a growing number of applications demanding highly sensitive photodetectors in the mid-infrared that does not require cooling. Thermal photodetectors, such as bolometers, have emerged as the technology of choice, because they do not need cooling.[2] Pyroelectric detectors offer major advantages in terms of cost and ease of operation over cooled photon detectors of long wavelength infrared radiation. Particularly, pyroelectric detectors are broadly reported due to their low power requirement, low manufacturing, fast response and relatively high sensitivity over a wide range of temperature and wide spectral bandwidth.[3] Nowadays, following a fast progress in terahertz (THz) and millimetre- wave (MMW) technologies, working on the improvement of the sensitivity of pyroelectric detection at longer wavelengths λ far beyond the IR range.[4] Pyroelectric detectors provide great advantages such as high sensitivity, uncooled or tunable operation, wide wavelength response, and low cost, which result in a variety of applications such as infrared sensing, thermal imaging, flame and fire alarms, gas monitoring.[5] Pyroelectricity is the ability of certain materials to produce surface charges due to a change in temperature. Those charges are a product of the spontaneous polarization observed in the opposite polar axis of the material, subjected to a temperature change. The pyroelectric effect is used for detecting the infrared radiation. Pyroelectric infrared detector offers the advantage of room temperature operation, wavelength independent sensitivity and low processing cost.

There are greater merits for thin film pyroelectric infrared detectors compared to bulk ceramic pyroelectric infrared detectors of similar materials. One of the merits is that the sensitivity of the thermal detectors can be improved by the reduced heat capacity of the detecting elements in thin film devices. Another merit is the potential to integrate the thin film detectors into a standard process of silicon chip fabrication.[6] If a pyroelectric material is exposed to radiation of power density W ($J^{-1}m^{-2}$) for a time, Δt, the radiation is absorbed onto the surface of the material which results in an increase in the temperature, ΔT (Fig. 1). It is assumed that all of the power absorbed in time Δt is rapidly distributed through the pyroelectric element volume, resulting in a uniform temperature increase. In this case, to simplify the process, heat losses from the pyroelectric are neglected. The increase in temperature is related to incident power density by Equation (1):[7]

$$\Delta T = \frac{W * \Delta t}{c_E * h} \quad (1)$$

$$= \frac{W * \Delta t}{c_p * \rho * h} \quad (2)$$

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where h is the pyroelectric thickness (m), c_p is the specific heat capacity ($Jkg^{-1}K^{-1}$), ρ is the density (kgm^{-3}), and c_E is the volume specific heat ($Jm^{-3}K^{-1}$). As the temperature of the pyroelectric material increases (*i.e.* dT/dt>0), there is a decrease in its level of the spontaneous polarization as dipoles within the material lose their orientation due to the thermal vibrations. This fall of the polarization level leads to a decrease in the number of unbound charges on the material surface.[8] The term pyroelectricity (from the Greek word Pyro-fire) was first introduced in 1824 by Brewster.[9,10]

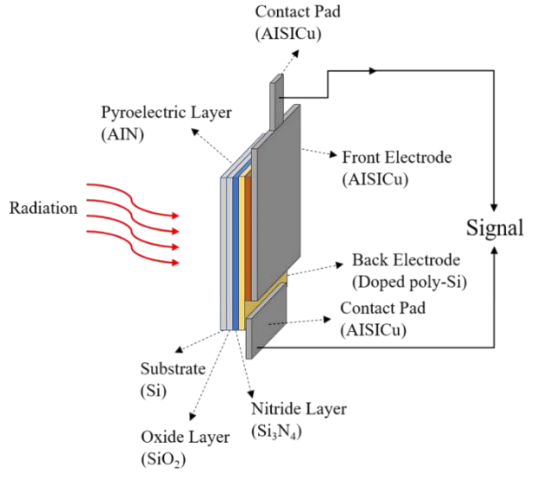

Fig. 1 Schematic representation of the pyroelectric detector.

The pyroelectricity is a coupled effect between temperature and electrical displacement (D, unit C/m^2) and represents the dependency of their change as shown by Equation (3):

$$dD = pd\theta, \quad (3)$$

where p is the pyroelectric coefficient with a unit C/m^2 K and it is defined as[11]

$$p = \left(\frac{dP_s}{d\theta}\right)_{T,E}, \quad (4)$$

where P_s is the spontaneous polarization and the constraints are constant electric field E and constant elastic stress T. Constant stress does not mean that the crystal is clamped, but completely free to expand or contract thermally.[12] There are two pyroelectric effects - primary and secondary. The primary pyroelectric effect means that a change in temperature causes a change in electrical displacement. The secondary pyroelectric effect is a result of crystal deformation - thermal expansion causes a strain that alters the electric displacement via a piezoelectric process. A different pyroelectric effect called tertiary pyroelectricity can occur by non-uniform heating causing non-uniform stresses that can result in polarization through a piezoelectric effect. In this case, the pyroelectric effect is the choice, which is about the spontaneous polarization in certain anisotropic solids as a result of temperature fluctuation.[13] This effect is of little interest now, although it is probably responsible for many unusual effects observed in the early works. The inverse pyroelectric effect, in which a change in an applied electric field produces a change in entropy and because of that - a change in temperature, is called electrocaloric effect.[12] The pyroelectric effect is connected with the ferroelectric (property of certain materials to have spontaneous polarization that can be reversed with an applied external electric field) and piezoelectric (property of certain materials to produce electric charge when subjected to a mechanical stress) effect. Pyroelectric materials have the potential to operate with a high thermodynamic efficiency and, compared to thermoelectrics, do not require bulky heat sinks to maintain the temperature gradient.[14] There are many piezoelectric materials, exhibiting the piezoelectric effect *i.e.* ability to generate electric charges in response to applied mechanical stress. Shifting of positive and negative charge centers in the material takes place when these materials are placed under mechanical stress. Most pyroelectric materials possess ferroelectric properties. This means that with an application of a suitable electric field, their polarization can be reversed. The polarization can be reduced to zero at a specific temperature called Curie temperature – T_c. Above this Curie temperature, the materials become paraelectric and they do not show a pyroelectric activity. However, it is possible to operate them as pyroelectric detectors with a temperature above T_c with the application of a suitable bias field if they operate near their T_c. This mode of operation is called dielectric bolometer and it requires additional equipment in order to operate.[9]

Monolithic pyroelectric detector has good properties such as fast response and high detectivity (D*), which shows applications in temperature sensing and pulse laser measurement.[15] Pyroelectric detectors utilize materials that possess a temperature dependent spontaneous polarization. This spontaneous polarization exists for temperature less than T_c of the material. Incident radiation heats up the detector, expanding the crystal lattice and changing the electric polarization (P) of the material.[11] Jon W. Stewart *et al.* experimentally and computationally demonstrated the pico- and nano- second dynamics of a room-temperature pyroelectric photodetector that is integrated with a metallic metasurface that acts as a perfect absorber and on-chip spectral filter.[16] Sergei A. Kuznetsov reports the development of a compact pyroelectric detector optimized for selective sensing of the MMW radiation within a narrow spectral band centered at 140 GHz (λ = 2.14 mm).[17]

2. Basic principles of the pyroelectric detector

A simple pyroelectric detector consists of a pyroelectric material with metal electrodes on the opposite face. These materials are oriented such that its polar axis is perpendicular to the electrode faces. To detect small changes, low noise high impedance amplifiers are necessary. Common arrangements is a simple JFET source.

3. Pyroelectric materials

There are five commonly used pyroelectric materials such as triglycine sulphate (TGS), strontium barium niobite (SBN),

lithium translate (LT) ceramic materials and polyvinylidene fluoride.

3.1. Triglycine sulphate crystals

TGS crystals are important because of their applications such as room temperature IR detector and imaging systems. These have the property to undergo second order ferroelectric phase transition at around 322 K.[18] TGS is the most sensitive and well-studied material. Researches have interest as it is easy to grow, has a high pyroelectric co-efficient, low dielectric permittivity, high voltage response and a high figure of merit.[19]

3.2. Strontium barium niobite

SBN is one such interesting ferroelectric materials with the TTB crystal structure which has applications in thermal/infrared detection and diverse device application including memory devices, surface acoustic wave devices *etc*.[20]

3.3. Lithium tantalate

LT gives a better performance than TGS due to its lower pyroelectric co-efficient and slightly higher relative permittivity. It has advantages of a very high T_c and it is insoluble in water. The detectors of these materials have responsivities, which are independent of temperature over a wide range.

3.4. Pyroelectric ceramics

These materials are relatively cheap to be manufactured in large areas using standard mixed oxide processes, they are both mechanically and chemically robust so that they can be readily processed into thin wafers for device fabrication.

Table 1 summarizes the properties of the most used pyroelectric materials in the infrared detectors and values of their sensor parameters at room temperature, where τ is thermal time constant of the pyroelectric detector, which is dependent on the thickness and temperature of the detector; R_v is voltage responsivity; NEP is noise equivalent power, which is ratio of the total noise to the voltage responsivity and is usually specified for a specific value of the wavelength, modulation frequency, bandwidth of the detector frequencies, temperature and cut-off frequency; D* is normalized detectivity and is written as D∗(T, f, 1), where T is the temperature in K, f is the frequency and 1 is the bandwidth for 1 Hz.[21]

Many materials that can be used for the production of infrared pyroelectric detectors exist. The choice of one is a difficult task because of many parameters involved in the calculation of the desired characteristics of the device that has to be designed, such as detector size, operating temperature, frequency of operation, and *etc*. The important characteristics that have to be noted in the choice of a material are low emissivity ε and loss, high pyroelectric coefficient and low thermal capacitance C_{th}. It is very difficult to point out a single material that can successfully satisfy all these requirements. Materials that have a high pyroelectric coefficient usually also have a high dielectric constant and vice versa, which determines the type of the amplifying electronic at the detector output. The pyroelectric materials have representatives to all three general categories well known for the semiconductors as well - single crystals, polycrystalline (ceramics) and amorphous (polymeric).

Although its pyroelectric coefficient is not the highest possible, according to Table 1, the most studied and used single crystals is the TGS – triglycine sulfate $(NH_2CH_2COOH)_3H_2SO_4$. Its performance parameters are excellent, such as high pyroelectric coefficient, low dielectric constant and low thermal conductivity. As to the disadvantages, it can be pointed out the poor long-term chemical and electrical stability and the low T_c making it difficult for processing with some of the microfabrication technologies. In spite of those drawbacks, TGS is often used for high performance detectors and vidicons. Several modifications of pure TGS were developed to overcome the low T_c issue. Aliane and arsenic acid doped materials (ATGSAs) show good performance having low dielectric losses and high pyroelectric coefficient.[22]

A material of lower performance than TGS, but possessing high chemical stability, low loss, good figure of merit, high

Table 1. Overview of the materials used in the state-of-the-art infrared detectors.

Material	Volume specific heat	Pyroelectric coefficient	Dielectric constant	τ [ms]	R_v [V/w]	NEP [W/Hz$^{1/2}$]	D* [cmHz$^{1/2}$/W]
TGS	1.70	3.5×10^{-8}	35	1390	7.05×10^{-14}	8.658×10^{-9}	1.41×10^{8}
$LiTaO_3$	3.19	1.9×10^{-8}	58	2600	1.23×10^{-14}	2.643×10^{-8}	4.63×10^{7}
LiNbO3	2.32	4.0×10^{-9}	75	1890	2.76×10^{-15}	1.623×10^{-7}	7.54×10^{6}
Li_2SO_4 xH_2O	0.82	1.0×10^{-8}	10	670	1.46×10^{-13}	8.658×10^{-9}	1.41×10^{8}
$BaTiO_3$	3.01	2.0×10^{-8}	4100	2460	1.94×10^{-16}	1.774×10^{-6}	6.9×10^{8}
$NaNO_2$	2.016	1.2×10^{-8}	8	1650	8.92×10^{-14}	5.773×10^{-9}	2.12×10^{8}
PVF_2	2.40	3.0×10^{-9}	11	1960	1.36×10^{-14}	3.175×10^{-8}	3.86×10^{7}
SBN	2.34	6.0×10^{-8}	400	1910	7.69×10^{-15}	5.771×10^{-8}	2.13×10^{7}
SbSi	2.378	2.6×10^{-7}	10^4	1940	1.31×10^{-15}	3.329×10^{-9}	3.68×10^{6}

curie temperature and good insolubility in water is the lithium tantalate - $LiTaO_3$. The material is widely used for single element detectors, although when used in low frequency devices, it can cause problems from thermally induced transient noise spikes. This material is not suitable for thermal imaging arrays and vidicons because of the low permittivity and high thermal conductivity.[9] Despite of the disadvantages and lower performance, $LiTaO_3$ is an affordable commercially and high-quality single crystals and can be produced by the Czochralski method. Ion slicing transferred single crystals thin films fabrication has been reported.[23] C. Ranacher *et al.*[1] describe various components of pyroelectric detector.

The next pyroelectric single crystal material is SBN - strontium barium niobate, consisting of complex elemental composition defined by the formula $Sr_{1-x}Ba_xNb_2O_6$, in which x can vary from 0,25 to 0,75. Depending on the composition, the ferroelectric transition can be tuned between 40 and 200 °C. This material is suitable for uncooled thermal imaging in near room temperature, because of its high dielectric constant.[24-25] The SBN can be produced by the Czochralski method, but high quality, defect free single crystals are difficult to grow. Pulse laser deposition with ablation is up-to-date technique that is applied for thin films SBN fabrication.[26]

Polymers based on polyvinylidene flouride (PVDF), polyvinyl fluoride (PVF) and on copolymers with trifluoroethylene (PVDF-TrFE) have a low pyroelectric coefficient, low dielectric constant and high losses, because they are amorphous.[27] They show lower performance characteristics compared to other pyroelectric materials but a comparison of often used sensors produced from them shows that they can have great advantages: they possess excellent mechanical properties, they have low thermal conductivity and low permittivity suitable for large area detectors, low heat conductivity and dielectric constant reduce the cross-talk between neighboring elements in multi-element detectors (between pyroelectric film and readout circuit); copolymer films can be easily deposited onto a substrate in post processing after readout circuit fabrication by spin, dip or spray coating of a copolymer solution.[28] Unlike conventional PVDF, when small amount of TrFE is introduced, the copolymer crystallizes directly into the ferroelectric phase. They can be easily cast from the melt of methyl ethyl ketone solution. That makes them suitable for direct deposition onto a silicon substrate for making arrays. The above mentioned polymers are affordable materials for use in the low-cost production of pyroelectric infrared sensors, arrays and vidicons.

A major part of the pyroelectric materials is the polycrystalline ferroelectric ceramics. This class of materials has great advantages over other types of pyroelectric materials: they are cheap to be manufactured in large areas using standard mixed-oxide processes; they possess good mechanical and chemical quality – and can be processed into thin wafers; they do not experience thermally induced noise spikes; they have high T_c; their performance parameters (pyroelectric coefficient, emissivity, resistivity, mechanical properties and thermal loss) can be modified with the use of selected dopant elements. A variety of solid solutions of PZ (lead zirconate - $PbZrO_3$) and PT (lead titanite - $PbTiO_3$), and similar oxides were developed to satisfy ferroelectric, piezoelectric, electro-optic and mainly – the pyroelectric requirements.[29]

Typical techniques for films deposition are sputtering and e-beam evaporation. Thin films can be grown textured or completely oriented in the case of epitaxy films. Improvements of the properties of ceramic materials which exist in bulk state are observed when grown in the form of thin film (like PZT, for example).[30] Pure $PbTiO_3$ is not preferable because of the high dielectric constant and difficulties to pole. Ceramics grown at around 1200º possess good properties - high emissivity and figure of merit, but for integrating ferroelectric thin films directly on Si substrates, it is important to grow the thin film at lower temperatures. The interconnect metallization on the chips should not be taken above 500º and this has to be the upper limit of the ferroelectric layer process temperature. For that reason, many techniques were developed, such as chemical solution deposition (CSD), sol-gel or metal-organic deposition (MOD), and metal-organic chemical vapor deposition (MOCVD).[31] Several groups of lead consisting pyroelectrics derivatives have been developed, like potassium thalium niobate - $KTa_xNb_{1-x}O_3$ (KTN); lead zinc niobate - $Pb(Zn_{1/3}Nb_{2/3})O_3$ (PZN); lead magnesium niobate - $Pb(Mg_{1/3}Nb_{2/3})O_3$ (PMN); and most recently - lead scandium tiantalate $Pb(Sc_{1/2}Ta_{1/2})O_3$ (PST). All those materials possess very high dielectric constants and high pyroelectric coefficients under the operating temperature and fields. Generally, they are desired for more specific demands, especially for very small area detector and large arrays of small elements. They are not suitable for low-cost applications.[32] Fig. 2 shows schematic representation of the pyroelectric sensor.

Suitable lead-free eco-friendly alternatives with comparable pyroelectric characteristics are the potassium niobate ($KNbO_3$) and barium strontium titanate ($BaSrTiO_3$), which can be grown by using the same approaches, as well as they can be nanostructured by anodic growth of aluminum oxide template filled with a potassium niobate to form nanowires for gaining sensitivity.[33]

The pyroelectric ceramics are a preferred choice for practical applications because of the stability of their properties over the normal operating temperature range. Their T_c is around 200 ºC and, because of that, they do not need an additional bias field to operate. Also the ceramics can be poled in any desired direction by the application of a suitable electrical field.[9] The typical resistivity of modified pyroelectric ceramics varies between 109 and 1011 Ω cm^2. This allows the developers to eliminate the presence of a gate bias resistor which generally is also around 1011 and 1012 Ωcm^2. This helps reduce the price for production in devices with many components (usually arrays

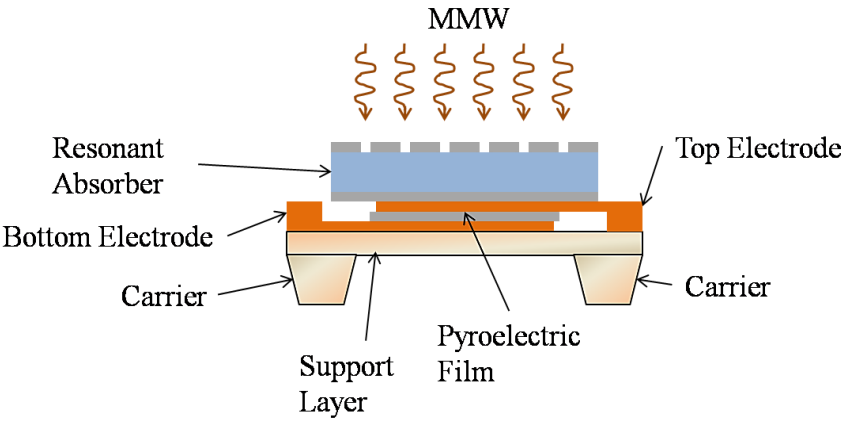

Fig. 2 Schematic representation of pyroelectric sensor.

of the detectors). Chi *et al.* described schematic drawing of the sandwich structure of porous (a) NKBT films and (b) NKBT/PLCT films.[34]

Zhang *et al.* measured the pyroelectric response of $BiFeO_3$ as a function of frequency at room temperature. From there, we showed that the pyroelectric response is increased by 100% with 15% lanthanum substitution.[35] Yaoa *et al.* reported the pyroelectric and piezoelectric coefficients at room temperature of 93, 87, 112, and 137 C/m^2 K, and 47, 52, 53, and 54 pC/N for their samples, respectively.[36] Yao *et al.* showed an anomaly in the temperature spectrum of the pyroelectric coefficient at around 70 to 80 °C. On the basis of their results, BFO may appear as a promising material for pyroelectric applications.[37] Fetisov *et al.* reported that multilayers of ferrite–PZT showed a weak pyroelectric coupling coefficient and found to have low resistivity and small ME coefficients.[38] Multilayers with a weak pyroelectric coupling are found to have low resistivity and small ME coefficients. The multilayers discussed here show an order of magnitude smaller ME coefficients compared to structurally superior samples.[39] Chi *et al.* described schematic drawing of the sandwich structure of porous (a) NKBT films and (b) NKBT/PLCT films, as is shown in Fig. 3.[34]

4. Conclusion and future work

Areas of engineering have tried to conquer various fields of application and other market. Infrared technology is conquering promising technologies, due to which it is gradually penetrating mass markets. These materials are extensively studied due to its giant magnetoelectric effect. In such structures, one usually measures alternative electric field produced due to magnetoelectric effect when the sample is placed in an external alternative magnetic field. Multilayers with that show weak pyroelectric coupling are found to have a low resistivity and small ME coefficients. It is very difficult to obtain a proper comparison between all pyroelectric materials because various parameters (like frequency and detector area) and environmental factors can affect the performance of the device. If a maximum detection ability is required, the element's area is of great importance, because it will affect the matching between the element capacitance and the amplifier capacitance. For large-area detectors, materials with a low dielectric constant are required. When high frequency is required, the pyroelectric polymers are suitable. For all other frequencies, TGA and lithium tantalite are the most suitable. For small area detectors (around 1 mm^2), no material is the best for all frequencies. For them, materials with high dielectric constants are better because of their better capacitance match between the element and the amplifier. For detectors with an intermediate area, the performance of all materials is more comparable. Other important factors when choosing a proper pyroelectric material are: environmental stability, production cost, availability, and manufacturing considerations.

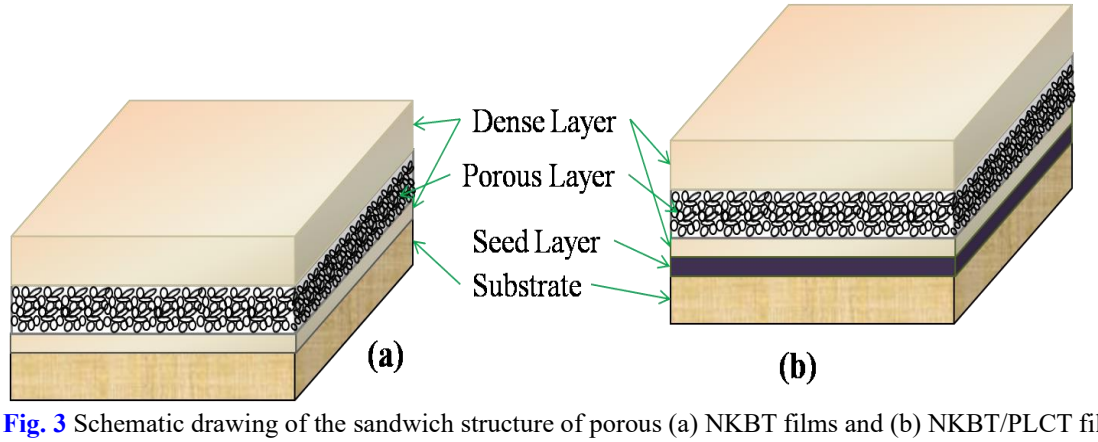

Fig. 3 Schematic drawing of the sandwich structure of porous (a) NKBT films and (b) NKBT/PLCT films.

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Conflict of interest
There are no conflicts to declare.

Supporting information
Not applicable.

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